Crucial role of the emitter–particle distance on the directivity of optical antennas

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We demonstrate that the reflecting properties of a single particle nanoantenna can be extremely sensitive to its distance from a quantum emitter at frequencies lower than the plasmon resonance. The phenomenon is shown to arise from rapid phase variations of the emitter field at short distances associated with a phase of the antenna particle polarizability lower than $\pi/4$. © 2011 Optical Society of America

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Optical antennas have been widely studied in this past decade for their ability to "focus" incident light into tiny volumes [1–3]. Reciprocally, the large amplification of the local electromagnetic density of states provoked by the antenna can increase the decay rates of nearby quantum emitters [3–10]. More recently, attention was focused on the ability of optical antennas to shape the radiation pattern of single quantum emitters to optimize light collection. With this goal in mind, different designs of unidirectional antennas have been introduced over the past four years [10–17]. Metallic nanoparticles can either reflect or attract the electric field radiated by the emitter. It is widely regarded that the reflecting/collecting nature of the particle depends solely on the polarizability of the nanoparticle: i.e., when the frequency of the emitter is larger than a particle's plasmon frequency, the particle will act as a reflector and when it is lower, the particle will act as a collector. The distance, d, between the exciting dipole and the center of the induced dipole is considerably smaller than the emission wavelength, $\lambda = 2\pi/k$, and one thus expects the phase of the particle polarizability to dominate the phase shift, kd, originating from the field propagation.

We study the directional property of a metallic particle of diameter $2a = 80\,\text{nm}$ made of silver (refractive index taken from [18]) transversely coupled with a single dipolar emitter ($\lambda_{\text{em}} = 600\,\text{nm}$ in vacuum) in a polymer-like embedding medium of index $n = 1.5$ [Fig. 1(a)]. For that purpose, we plot first the reflected power efficiency defined as the ratio of the Poynting vector integrated over the left half-space ($\cos\varphi < 0$) with respect to its integration over all directions, as a function of the separation distance between the two dipoles. The calculations were performed using the generalized Mie theory with a multipole order $n_{\max} = 30$. We recently improved the formalism to analytically calculate the flow of the Poynting vector over a spherical surface [19], which significantly decreases the calculation time of the decay rates. The redshift of the emission frequency with respect to the plasmon resonance (around 500 nm; see inset in Fig. 2) predicts a collector behavior as observed with a minimum of 28% reflected power when $d = 78\,\text{nm}$. This collector behavior is lost entirely at a distance $d = 59\,\text{nm}$ where the particle neither reflects nor collects the emission (50% reflected/collected power). But even more remarkably, at a distance of $d = 49\,\text{nm}$ (i.e., 9 nm from the metal surface), it acts as a good reflector (91% reflected power). We note that the reflection efficiency is well predicted by a dipolar approximation, even when the emitter approaches the metallic surface to within a few nanometers. The contribution of higher order multipoles is significant when the emitter is 20 nm or less away from the metallic surface. However, the dissipation of multipolar modes mostly occurs by the Joule effect, with minimal far-field radiation compared to the dipolar component [7]. Consequently, the dipolar model introduces only a 2% relative error for $d = 49\,\text{nm}$ and this error becomes negligible as soon as $d > 59\,\text{nm}$. This is not the case for the fluorescence decay rates that clearly require higher multipolar terms for $d < 60\,\text{nm}$ with an order of magnitude discrepancy at $d = 45\,\text{nm}$. The radiation patterns reconstructed with the rigorous generalized Mie theory, without any dipolar approximation (multipole order

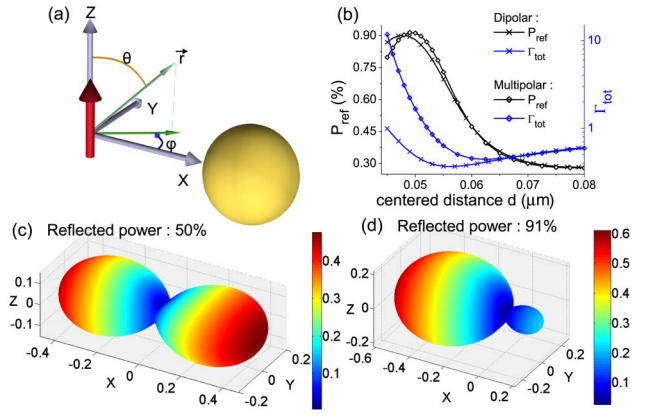

Fig. 1. (Color online) (a) An electric dipole emitter oriented along the z axis is coupled to a sphere whose center is at a distance d along the x axis. (b) Reflected power efficiency P_{ref} (left scale, top full lines) and total decay rates Γ_{tot} (right scale, logarithmic scale, bottom full lines) with respect to the separation d: crosses, dipolar approximation; squares, multipolar calculations. (c), (d) Radiation patterns with separation distances, equal to (c) $d = 59\,\text{nm}$ and (d) $d = 49\,\text{nm}$.

$n_{\max} = 30$), respectively for $d = 49\,\text{nm}$ and $d = 59\,\text{nm}$ [Figs. 1(c) and 1(d)] confirm the drastic modification of the radiation directionality with a minute modification of only 10 nm in the distance between the emitter and the metallic surface.

These results are of crucial importance for nanoantenna applications, and we now aim to analytically explain the physical mechanisms involved in this extreme position sensitivity of the emission directivity when the emitter is separated by a few nanometers from the metallic surface. Therefore, we focus on the interaction between a single metallic particle and a dipolar emitter, and we only consider the transverse coupling geometry that provides significant emission directionality. In this case, fluorescence decay rates are weakly enhanced compared to longitudinal interactions [7]. However, more complex antenna geometries can be designed to combine rate enhancements and directionality by longitudinally coupling the quantum emitter to a nanogap antenna ("super emitter") [4,13,16] or the feed element of a Yagi–Uda antenna [15]. In all these examples, emission directionality is governed by phase differences between transversely coupled dipoles, as in this demonstrative study.

We consider a unit dipole placed at origin, oriented along the z axis ($\mathbf{p}_{\text{em}} = \hat{\mathbf{z}}$) and a spherical scatterer whose center is located at a distance d along the x axis. The excitation field produced by a dipole emitter at this position is written [20]:

$$\begin{aligned}\mathbf{E}(d) &= \frac{e^{ikd}}{4\pi\varepsilon_m\epsilon_0 d^3}[k^2d^2(\hat{\mathbf{x}}\times\mathbf{p}_{\text{em}})\times\hat{\mathbf{x}} \\ &\quad + (1-ikd)(3(\hat{\mathbf{x}}\cdot\mathbf{p}_{\text{em}})\hat{\mathbf{x}} - \mathbf{p}_{\text{em}})] \\ &= -\frac{e^{ikd}}{4\pi\varepsilon_m\epsilon_0 d^3}(1-ikd-k^2d^2)\hat{\mathbf{z}}, \qquad (1)\end{aligned}$$

and the induced dipolar moment in the sphere is thus

$$\mathbf{p}_{\text{in}} = -\alpha\frac{e^{ikd}}{4\pi d^3}(1-ikd-k^2d^2)\hat{\mathbf{z}}.$$

The polarizability, α, of a spherical scatterer is calculated analytically using the Mie theory [21]. The dipolar model studied here is not limited to spheres but can be extended to other geometries as long as the dipole plasmon mode dominates. This is particularly true for rod-shaped particles (often used in Yagi–Uda antennas) for which the longitudinal dipolar mode is strongly redshifted with respect to higher multipoles. In order to quantify the influence of the induced dipole moment of a metallic nanoparticle on the emission pattern of the emitter, we calculate for $\cos\varphi > 0$ and $r \gg d$, the Poynting vector of the field emitted by two transversely coupled dipoles (denoted $\mathbf{p}_1$ and $\mathbf{p}_2$), and then we add the Poynting vector symmetric with respect to the origin [16]:

$$\begin{aligned}\Delta\mathbf{P}(r,\theta,\varphi) &= \mathbf{P}(r,\theta,\varphi) + \mathbf{P}(r,\pi-\theta,\pi+\varphi) \\ &= \frac{\omega^3 k|p_1||p_2|}{8\pi^2\epsilon_0 c^2 r^2}\{\sin\phi\sin[kd\sin\theta\cos\varphi]\}\sin^2\theta\mathbf{e}_{\mathbf{r}},\end{aligned}$$

where $\phi = \arg(p_1/p_2)$ is the relative phase of the two dipoles. This expression confirms that for small kd, the directivity is directly linked to the sign of $\sin(\phi)$, i.e., to the capacitive or inductive behavior of the dipolar metallic particle [11]. For emissions along the x axis, $\sin\theta = \cos\varphi = 1$, and the last expression simplifies to

$$\Delta\mathbf{P}(x,d) = \frac{\omega^3 k|p_1||p_2|}{8\pi^2\epsilon_0 c^2 x^2}\sin\phi\sin(kd)\hat{\mathbf{x}}.$$

If the relative phase, ϕ, of the two dipoles was simply equal to kd as far-field reasoning would suggest, this last expression would predict that the collector/reflector behavior of the nanoparticle oscillates with separation distance as $\sin^2(kd)$. In this case, the strong changes in emission directivity for minute distance variations observed in Fig. 1 would remain unexplained. However, the phase difference, ϕ, between the emitter ($\mathbf{p}_{\text{em}}\cdot\hat{\mathbf{z}} = 1$) and the induced dipole ($\mathbf{p}_{\text{in}}\cdot\hat{\mathbf{z}} = -\alpha\frac{e^{ikd}}{4\pi d^3}(1-ikd-k^2d^2)$) is a nonlinear function of kd:

$$\phi(kd) \equiv \arg\left(\frac{\mathbf{p}_{\text{in}}\cdot\hat{\mathbf{z}}}{\mathbf{p}_{\text{em}}\cdot\hat{\mathbf{z}}}\right) = \arg[-\alpha e^{ikd}(1-ikd-k^2d^2)].$$

The phase due to the distance between the dipoles is

$$\phi_d \equiv \phi - \arg(\alpha) = \arg[-e^{ikd}(1-ikd-k^2d^2)]. \qquad (2)$$

Two different terms determine ϕ_d: the well-known propagative or "far-field" phase term, kd, in the exponential, and the phase of the dipolar field term (i.e., the argument of $k^2d^2 + ikd - 1$). The plots in Fig. 2 of ϕ_d, together with the dipolar (dashed curve) and far-field (circles) contributions, as a function of the dimensionless parameter, kd, show that for small distances, i.e., $kd \le \pi/4$, the distance dependent phase difference is dominated by the dipolar contribution. When $kd \to 0$, $\phi_d \to \pi$ due to the minus sign in Eq. (2). This is the expected phase difference for a near-field electrostatic interaction dominated by the $1/d^3$ term in Eq. (1). Consequently, $\sin\phi \to -\sin[\arg(\alpha)]$,

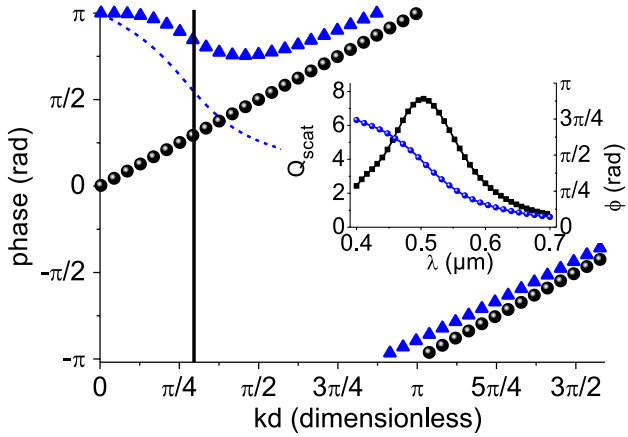

Fig. 2. (Color online) Triangles, ϕ_d for an emitter transversely coupled to a spherical scatterer as a function of kd; dashed line, dipolar field contribution; circles, far-field term contribution. Vertical line marks the $kd = 0.93$ abscissa. Inset (circles, left scale), polarizability phase, $\text{Arg}(\alpha)$, and (squares, right scale) scattering efficiency, $Q_{\text{scat}} = \sigma_s/\pi a^2$, of a 80 nm diameter silver sphere, both as a function of the vacuum wavelength.

which is negative (see inset in Fig. 2) and thus $\Delta\mathbf{P} < 0$, meaning that the particle behaves as a reflector.

Let us now discuss the situation where kd is small but not vanishing. The phase contribution from the dipolar field term decreases rapidly with respect to kd while the far-field term increases linearly. Consequently, for small distances, ϕ_d decreases with respect to kd. One can see in Fig. 2 that a minimum of $\phi_d \simeq 3\pi/4$ occurs at $kd = \sqrt{2}$. This means that the particle can behave as a collector, $\Delta\mathbf{P} > 0$, provided that $\phi_\alpha \equiv \arg(\alpha) < \pi/4$. This condition is generally fulfilled for wavelengths significantly larger than the particle plasmon resonance frequency. This combination ($kd \simeq \sqrt{2}$, $\phi_\alpha < \pi/4$) is the only possibility for a transversely coupled metallic particle to act as a collector at "small" distances ($kd < 3\pi/4$). In other words, for a metallic particle with $\arg(\alpha) > \pi/4$ at small distances ($kd < 3\pi/4$), the condition $\Delta\mathbf{P} = 0$ cannot be achieved and thus the directional property cannot be reversed by varying the phase of the dipolar term (the particle can only be a reflector). In such cases, tuning the behavior of a single particle from reflecting to collecting requires working with larger separations for which the far-field term kd of Eq. (2) dominates. In practice, Fig. 2 demonstrates a clear transition between the electrostatic approximation ($kd \to 0$, $\phi_d = \pi$) and the far-field approximation (circles in Fig. 2). In this transition region, the dipolar field phase term strongly influences the antenna behavior around $kd = \pi/4$, where its slope versus kd is highest. For this distance range, the emitter–particle coupling depends strongly on the ik/d^2 term of Eq. (1) and is thus dominated by a phase-dependent dipole–dipole interaction.

Using this dipolar model, we are able to explain the surprising phenomena shown in Fig. 1. With $\lambda_{\mathrm{em}} = 600\,\mathrm{nm}$ and $d = 59\,\mathrm{nm}$, we have $kd \cong 0.93$, which gives $\phi_d = 2.64\,\mathrm{rad}$ (see the vertical line in Fig. 2). The phase of the polarizability of the silver particle at this wavelength is 0.50 rad (see inset). The total relative phase between the exciting and induced dipoles is then $\phi = 3.14\,\mathrm{rad} \cong \pi\,\mathrm{rad}$, which results in $\sin\phi \cong 0$, i.e., a symmetric emission pattern. Starting from $d = 59\,\mathrm{nm}$, when d decreases, ϕ_d increases, resulting in $\sin\phi < 0$, thus explaining the strong reflecting behavior of the sphere placed at a distance of 49 nm. Respectively, a slight increase of d results in $\sin\phi > 0$ and a collecting behavior for the sphere.

We have shown that one can finely tune the directional behavior of optical antennas by engineering the relative phases of the emitting and induced dipoles. For emission wavelengths larger than the particle plasmon frequencies, the phase of the particle polarizability can drop below $\pi/4$ and the radiation pattern becomes strongly dependent on the emitter position at a $\lambda/60$ scale. We have shown that this phenomenon is due to rapid variations of the phase of the dipole–dipole coupling term when the ik/d^2 term of the electric field expression is nonnegligible. This result provides new insights into the conception of ultracompact optical antennas and should inspire experimental investigations to fully unravel the dephasing effects that govern the interaction between single emitters and metallic nanostructures.

References

1. S. S. Acimovic, M. P. Kreuzer, M. U. Gonzalez, and R. Quidant, ACS Nano **3**, 1231 (2009).
2. A. Devilez, B. Stout, and N. Bonod, Phys. Rev. B **81**, 245128 (2010).
3. L. Novotny and N. van Hulst, Nat. Photon. **5**, 83 (2011).
4. J. N. Farahani, D. W. Pohl, H.-J. Eisler, and B. Hecht, Phys. Rev. Lett. **95**, 017402 (2005).
5. P. Anger, P. Bharadwaj, and L. Novotny, Phys. Rev. Lett. **96**, 113002 (2006).
6. S. Kuhn, U. Hakanson, L. Rogobete, and V. Sandoghdar, Phys. Rev. Lett. **97**, 017402 (2006).
7. H. Mertens, A. F. Koenderink, and A. Polman, Phys. Rev. B **76**, 115123 (2007).
8. L. Rogobete, F. Kaminski, M. Agio, and V. Sandoghdar, Opt. Lett. **32**, 1623 (2007).
9. A. Kinkhabwala, Z. Yu, S. Fan, Y. Avlasevich, K. Mullen, and W. E. Moerner, Nat. Photon. **3**, 654 (2009).
10. H. Aouani, O. Mahboub, N. Bonod, E. Devaux, E. Popov, H. Rigneault, T. W. Ebbesen, and J. Wenger, Nano Lett. **11**, 637 (2011).
11. J. Li, A. Salandrino, and N. Engheta, Phys. Rev. B **76**, 245403 (2007).
12. T. H. Taminiau, F. D. Stefani, F. B. Segerink, and N. F. van Hulst, Nat. Photon. **2**, 234 (2008).
13. A. F. Koenderink, Nano Lett. **9**, 4228 (2009).
14. A. Devilez, B. Stout, and N. Bonod, ACS Nano **4**, 3390 (2010).
15. A. Curto, G. Volpe, T. Taminiau, M. Kreuzer, R. Quidant, and N. F. van Hulst, Science **329**, 930 (2010).
16. N. Bonod, A. Devilez, B. Rolly, S. Bidault, and B. Stout, Phys. Rev. B **82**, 115429 (2010).
17. R. Esteban, T. V. Teperik, and J. J. Greffet, Phys. Rev. Lett. **104**, 026802 (2010).
18. E. Palik and G. Ghosh, *Handbook of Optical Constants of Solids* (Academic, 1998).
19. B. Stout, A. Devilez, B. Rolly, and N. Bonod, J. Opt. Soc. Am. B **28**, 1213 (2011).
20. L. Novotny and B. Hecht, *Principles of Nano-Optics* (Cambridge Univ. Press, 2006), pp. 250–303.
21. C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (Wiley, 1983).